

## PATENT COOPERATION TREATY

## PCT

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## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)


10/529528

Applicant's or agent's file reference 62820A		<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/US 03/33192	International filing date (day/month/year) 17.10.2003	Priority date (day/month/year) 17.10.2002	
International Patent Classification (IPC) or both national classification and IPC C08L23/08			
Applicant DOW GLOBAL TECHNOLOGIES INC. et al.			

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 5 sheets, including this cover sheet.
  - ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 6 sheets.

3. This report contains indications relating to the following items:
  - I ☒ Basis of the opinion
  - II ☐ Priority
  - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
  - IV ☐ Lack of unity of invention
  - V ☒ Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
  - VI ☐ Certain documents cited
  - VII ☐ Certain defects in the international application
  - VIII ☐ Certain observations on the international application

Date of submission of the demand  14.05.2004	Date of completion of this report  11.01.2005
Name and mailing address of the international preliminary examining authority:   European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized Officer  Frison, C  Telephone No. +49 89 2399-8519



**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. **PCT/US 03/33192**

**I. Basis of the report**

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17):*

**Description, Pages**

1-7, 9, 10, 12-15, 17-23 as originally filed  
8, 11, 16 received on 28.10.2004 with letter of 28.10.2004

**Claims, Numbers**

1-9 received on 28.10.2004 with letter of 28.10.2004

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).  
☐ the language of publication of the international application (under Rule 48.3(b)).  
☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.  
☐ filed together with the international application in computer readable form.  
☐ furnished subsequently to this Authority in written form.  
☐ furnished subsequently to this Authority in computer readable form.  
☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.  
☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:  
☐ the claims, Nos.:  
☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

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**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability;  
citations and explanations supporting such statement**

**1. Statement**

Novelty (N)	Yes: Claims	1-9
	No: Claims	
Inventive step (IS)	Yes: Claims	1-9
	No: Claims	
Industrial applicability (IA)	Yes: Claims	1-9
	No: Claims	

**2. Citations and explanations**

**see separate sheet**

Concerning point V

1. Art. 33(2) PCT

*claim 1*

It is now clear from claim 1 that the polymer composition comprises A and B (amount of B not equal to 0). From the relationship between the Brookfield viscosity (at 177°C) and the weight average molecular weight (Mw), it can be calculated that the base polymer has a Mw of not more than ca. 37,000.

D1 discloses an ethylene/propylene diene terpolymer (Vistalon) having a total crystallinity of less than 1% but has a weight average molecular weight greater than 37,000 (360,400: see Table III). As mentioned in the written opinion dated 28.07.2004, a further composition is described in Table IV, said composition comprising an ethylene/octene copolymer having a crystallinity of more than 30%.

The non-crystalline EPDM polymers disclosed in document D2 (see Table II) have a weight average molecular weights of 232,300 and 243,500.

Document D3 discloses a thermoplastic resin composition comprising a propylene/alpha-olefin random copolymer modified by grafting of unsaturated carboxylic acid or their derivatives, said copolymer having a crystallinity of 0-30%, and a high amount of inorganic filler. There is no indication in the Derwent abstract, nor in the Chemical Abstract that the copolymer has a low viscosity has claimed in claim 1. The benefit of the doubt is given to the applicant. However, it can not be ruled out that in the European Phase, solid arguments proving that the polymer exemplified in document D3 has a higher viscosity than in the present application could be requested by the examining division.

At present, the subject-matter of claim 1 is considered to be novel over D1-D3

2. Art 33(3) PCT

Document D1, which is considered to represent the most relevant state of the art, discloses a polymer composition from which the subject-matter of claim 1 differs in that the base polymer comprised in the composition has a lower viscosity. The technical effect linked to this distinguishing feature is a processability at lower torque and lower melt temperature (see Table 2). The objective problem to solve can therefore be formulated as to provide

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EXAMINATION REPORT - SEPARATE SHEET**

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a polymer composition with high loading of filler, which can be processed with lower energy. None of the prior art suggest that this problem can be solved by using a low viscosity polymer with a low crystallinity. Besides, none of the prior art aims at solving this problem.

An inventive step may therefore be acknowledged.

*Claim 9*

Claim 9 could be objected as lacking clarity in the European Phase because the subject-matter of claim 9 can be defined more precisely without unduly restricting the scope of the claim (see guideline, CIII, 4.7). Indeed, the result (to obtain a high calcium carbonate loaded mixture having a viscosity greater than  $1.3 \times 10^4$  poise) can be achieved by using a base polymer having the characteristics expressed in claim 1. Otherwise, the Examining Division does not understand the meaning of present claim 9.

WO 2004/035680

PCT/US2003/033192

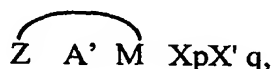
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REPLACED BY  
ART 34 AMDT

methylenetitanium dichloro, (tertbutylamido)diphenyl(tetramethyl- $\eta^5$ -cyclopentadienyl)-silane, zirconium dibenzyl, (benzylamido)dimethyl-(tetramethyl- $\eta^5$ -cyclopentadienyl) ilanetitaniumdichloride, and phenylphosphido)dimethyl(tetramethyl- $\eta^5$ -cyclopentadienyl) silane

zirconium dibenzyl. Another suitable class of catalysts is substituted indenyl containing metal complexes as disclosed in U.S. Patents No. 5,965,756 and No. 6,015,868. Other preferred catalysts are disclosed in US Patent 5,616,664 and copending applications: U.S. Application Serial No. 09/230,185; and No. 09/715,380, and U.S. Provisional Application Serial No. 60/215,456; No. 60/170,175, and No. 60/393,862. These catalysts tend to have a higher molecular weight capability.

One class of the above catalysts is the indenyl containing metal wherein:



Formula IV

M is titanium, zirconium or hafnium in the +2, +3 or +4 formal oxidation state;

A' is a substituted indenyl group substituted in at least the 2 or 3 position with a group selected from hydrocarbyl, fluoro-substituted hydrocarbyl, hydrocarbyloxy-substituted hydrocarbyl, dialkylamino- substituted hydrocarbyl, silyl, germyl and mixtures thereof, the group containing up to 40 non-hydrogen atoms, and the A' further being covalently bonded to M by means of a divalent Z group; Z is a divalent moiety bound to both A' and M via  $\sigma$ -bonds, the Z comprising boron, or a member of Group 14 of the Periodic Table of the Elements, and also comprising nitrogen, phosphorus, sulfur or oxygen; X is an anionic or dianionic ligand group having up to 60 atoms exclusive of the class of ligands that are cyclic, delocalized,  $\pi$ -bound ligand groups; X' independently each occurrence is a neutral Lewis base, having up to 20 atoms; p is 0, 1 or 2, and is two less than the formal oxidation state of M, with the proviso that when X is a dianionic ligand group, p is 1; and q is 0, 1 or 2.

The above complexes may exist as isolated crystals optionally in pure form or as a mixture with other complexes, in the form of a solvated adduct, optionally in a solvent, especially an organic liquid, as well as in the form of a dimer or chelated derivative thereof, wherein the chelating agent is an organic material, preferably a neutral Lewis base, especially a trihydrocarbylamine, trihydrocarbylphosphine, or halogenated derivative thereof.

US Patent 5,621,127; US Patent 5,703,257; US Patent 5,728,855; US Patent 5,731,253; US Patent 5,710,224; US Patent 5,883,204; US Patent 5,504,049; US Patent 5,962,714; US Patent 5,965,677; US Patent 5,427,991; WO 93/21238, published October 28, 1993; WO 94/03506, published February 17, 1994; WO 93/21242, published October 28, 1993; WO 94/00500, published January 6, 1994; WO 96/00244, published January 4, 1996; WO 98/50392, published November 12, 1998; Wang, et al., Organometallics 1998, 17, 3149-3151; Younkin, et al., Science 2000, 287, 460-462, Chen and Marks, Chem. Rev. 2000, 100, 1391-1434, Alt and Koppl, Chem. Rev. 2000, 100, 1205-1221; Resconi, et al., Chem. Rev. 2000, 100, 1253-1345; Ittel, et al., ChemRev. 2000, 100, 1169-1203; Coates, Chem. Rev., 2000, 100, 1223-1251; WO 96/13530, published May 9, 1996. Also useful are those catalysts, cocatalysts, and catalyst systems disclosed in USSN 09/230,185, filed January 15, 1999; US Patent 5,965,756; US 6,150,297; USSN 09/715,380, filed November 17, 2000. In addition, methods for preparing the aforementioned catalysts are described, for example, in U.S. Patent No. 6,015,868.

The above-described catalysts may be rendered catalytically active by combination with an activating cocatalyst or by use of an activating technique. Suitable activating cocatalysts for use herein include, but are not limited to, polymeric or oligomeric alumoxanes, especially methylalumoxane, triisobutyl aluminum modified methylalumoxane, or isobutylalumoxane; neutral Lewis acids, such as C<sub>1-30</sub> hydrocarbyl substituted Group 13 compounds, especially tri(hydrocarbyl)aluminum- or tri(hydrocarbyl)boron compounds and halogenated (including perhalogenated) derivatives thereof, having from 1 to 30 carbons in each hydrocarbyl or halogenated hydrocarbyl group, more especially perfluorinated tri(aryl)boron and perfluorinated tri(aryl)aluminum compounds, mixtures of fluoro-substituted(aryl)boron compounds with alkyl-containing aluminum compounds, especially mixtures of tris(pentafluorophenyl)borane with trialkylaluminum or mixtures of tris(pentafluorophenyl)borane with alkylalumoxanes, more especially mixtures of tris(pentafluorophenyl)borane with methylalumoxane and mixtures of tris(pentafluorophenyl)borane with methylalumoxane modified with a percentage of higher alkyl groups (MMAO), and most especially tris(pentafluorophenyl)borane and tris(pentafluorophenyl)-aluminum; non-polymeric, compatible, non-coordinating, ion forming compounds (including the use of such compounds under oxidizing conditions), especially the use of ammonium-, phosphonium-, oxonium-, carbonium-, silylium- or sulfonium- salts of compatible, non-coordinating anions, or ferrocenium salts of compatible, non-coordinating anions; bulk electrolysis and combinations of the foregoing activating cocatalysts and techniques. The foregoing activating cocatalysts and activating techniques have been previously taught with respect to different metal complexes in the following references: EP-A-277,003, US-A-5,153,157,

The solvent will be present in an amount sufficient to prevent phase separation in the reactor. As the solvent functions to absorb heat, less solvent leads to a less adiabatic reactor. The solvent:ethylene ratio (weight basis) will typically be from 2.5 : 1 to 12 : 1, beyond which point catalyst efficiency suffers. The most typical solvent:ethylene ratio (weight basis) is in the range of from 5 : 1 to 10 : 1.

The polymerization may be carried out as a batchwise or a continuous polymerization process, with continuous solution polymerizations processes being most preferred for the preparation of the liquid low molecular weight polymers of the invention. In a continuous process, ethylene, comonomer, and optionally solvent and diene are continuously supplied to the reaction zone and polymer product continuously removed therefrom.

The homogeneous, low crystallinity, low viscosity ethylene and/or alpha olefin homopolymers and interpolymers used as the base polymer for the filled compositions of the present invention may be polymerized in a first reactor, with a second polymer (of higher molecular weight and/or of different density, and/or which is heterogeneous) being polymerized in a second reactor which is connected in series or in parallel to that in which the liquid low molecular weight polymer is produced, to prepare in-reactor polymer blends having desirable properties. An example of a dual reactor process which may be adapted in accordance with the teachings of this disclosure to prepare blends wherein at least one component is the homogeneous liquid low molecular weight ethylene/alpha-olefin polymer of this invention, is disclosed in WO 94/00500, (equivalent to U. S. Serial Number 07/904,770), as well as USSN 08/10958, filed January 29, 1993.

The polymer may also be prepared using a dual catalyst system in either a single, dual or multiple reactor configuration as disclosed in copending US Patent Application Serial No.60/504,412 filed on September 19, 2003 in the Teresa Karjala and Brian Kolthammer.

The homogeneous low crystallinity, low viscosity ethylene and/or alpha olefin homopolymers or interpolymers used as the base polymer for the filled compositions of the present invention may also be blended with one or more other polymers prior to mixing with the additive. Such other blend polymers include but are not limited to, styrene block copolymers, rubbers, linear low density polyethylene (LLDPE), high density polyethylene (HDPE), low density polyethylene (LDPE), ethylene/vinyl acetate (EVA) copolymer, ethylene-carboxylic acid copolymers (EAA), ethylene acrylate copolymers, polybutylene, polybutadiene, nylons, polycarbonates, polyesters, polypropylene, ethylene-propylene interpolymers such as ethylene-propylene rubber, ethylene-propylene-diene monomer rubbers, chlorinated polyethylene, thermoplastic vulcanates, ethylene ethylacrylate polymers (EEA), ethylene styrene interpolymers



REPLACED BY  
ART 34 AMDTWHAT IS CLAIMED IS:

1. A polymer composition comprising;

A) greater than or equal to 40 percent by weight (based on the combined weights of

5 Component A and B) of one or more fillers; and

B) less than 60 percent by weight (based on the combined weights of Component A and B) of one or more base polymers;

wherein said base polymer has

1) a total crystallinity of from 0 to 30 percent; and

10 2) a Brookfield viscosity of from 500 to 50,000 cP measured at 350°F.

2. The polymer composition of Claim 1 wherein;

A) said one or more fillers, Component A, is present in an amount of greater than or equal to 60 percent by weight (based on the combined weights of Component A and B); and

15 B) said one or more base polymers, Component B, is present in an amount of less than 40 percent by weight (based on the combined weights of Component A and B); and

wherein said base polymer has

1) a total crystallinity of from 3 to 25 percent; and

2) a Brookfield viscosity of from 2,000 to 30,000 cP measured at 350°F.

20

3. The polymer composition of Claim 1 wherein;

A) said one or more fillers, Component A, is present in an amount of greater than or equal to 80 percent by weight (based on the combined weights of Component A and B); and

25 B) said one or more base polymers, Component B, is present in an amount of less than 20 percent by weight (based on the combined weights of Component A and B); and

wherein said base polymer has

1) a total crystallinity of from 5 to 20 percent; and

2) a Brookfield viscosity of from 5,000 to 20,000 cP measured at 350°F.

**REPLACED BY  
ART 34 AMDT**

4. The polymer composition of Claim 1 wherein;

A) said one or more fillers, Component A, is selected from the group consisting of glass fibers, talc, calcium carbonate, alumina trihydrate, glass fibers, marble dust, cement dust, clay, feldspar, silica or glass, fumed silica, alumina, magnesium oxide, magnesium hydroxide, antimony oxide, zinc oxide, barium sulfate, aluminum silicate, calcium silicate, titanium dioxide, titanates, glass microspheres or chalk, hindered phenolics, phosphites, light stabilizers,; plasticizers; tackifiers; waxes; processing aids; stearic acid or a metal salt thereof; crosslinking agents; colorants or pigments; carbon black; graphite; carbon fibers; and blowing agents; and any and all combinations thereof; and

B) said one or more base polymers, Component B, is a homogenous ethylene/C<sub>3</sub>-C<sub>20</sub> alpha-olefin interpolymer or a C<sub>3</sub>-C<sub>20</sub> alpha olefin homopolymer or interpolymer.

5. The polymer composition of Claim 4 wherein;

A) said one or more fillers, Component A, is carbon black; alumina trihydrate, calcium carbonate or any and all combinations thereof; and

B) said one or more base polymers, Component B, is a homogenous ethylene/propylene or ethylene/octene-1 copolymer or polypropylene.

6. The polymer composition of Claim 1 wherein

i) when calcium carbonate is said filler, the highest ultimate filler loading is greater than 90 weight percent; or

ii) when alumina trihydrate is said filler, the highest ultimate filler loading is greater than 80 weight percent.

7. A polymer composition comprising a filler and polymer, wherein when said polymer is mixed with calcium carbonate at a concentration of 89 weight percent the viscosity of the resulting mixture (when measured on a Rheometrics RMS-800 with 25 mm parallel plates at frequency of 100 rad/s at 230 °C in a nitrogen purge) is greater than  $1.3 \times 10^4$  poise.

**AMENDED CLAIMS**

received by the International Bureau on 16 March 2004 (16.03.04).  
original claims 1, 4, and 7 are replaced by amended claims 1, 4 and 7 (2 pages)]

REPLACED BY  
ART 34/ADT

1. A polymer composition comprising;

A) greater than or equal to 40 percent by weight (based on the combined weights of Component A and B) of one or more fillers; and

5 B) less than 60 percent by weight (based on the combined weights of Component A and B) of one or more base polymers;

wherein said one or more base polymers is a homogenous ethylene/C<sub>3</sub>-C<sub>20</sub> alpha-olefin interpolymer or a C<sub>3</sub>-C<sub>20</sub> homopolymer or interpolymer, and has

1) a total crystallinity of from 0 to 30 percent; and

10 2) a Brookfield viscosity of from 500 to 50,000 cP measured at 350°F.

2. The polymer composition of Claim 1 wherein;

A) said one or more fillers, Component A, is present in an amount of greater than or equal to 60 percent by weight (based on the combined weights of Component A and B); and

15 B) said one or more base polymers, Component B, is present in an amount of less than 40 percent by weight (based on the combined weights of Component A and B); and

wherein said base polymer has

1) a total crystallinity of from 3 to 25 percent; and

2) a Brookfield viscosity of from 2,000 to 30,000 cP measured at 350°F.

20

3. The polymer composition of Claim 1 wherein;

A) said one or more fillers, Component A, is present in an amount of greater than or equal to 80 percent by weight (based on the combined weights of Component A and B); and

25 B) said one or more base polymers, Component B, is present in an amount of less than 20 percent by weight (based on the combined weights of Component A and B); and

wherein said base polymer has

1) a total crystallinity of from 5 to 20 percent; and

2) a Brookfield viscosity of from 5,000 to 20,000 cP measured at 350°F.

**REPLACED BY  
ART 34 AMDT**

4. The polymer composition of Claim 1 wherein;

A) said one or more fillers, Component A, is selected from the group consisting of glass fibers, talc, calcium carbonate, alumina trihydrate, glass fibers, marble dust, cement dust, clay, feldspar, silica or glass, fumed silica, alumina, magnesium oxide, magnesium hydroxide, antimony oxide, zinc oxide, barium sulfate, aluminum silicate, calcium silicate, titanium dioxide, titanates, glass microspheres or chalk, hindered phenolics, phosphites, light stabilizers,; plasticizers; tackifiers; waxes; processing aids; stearic acid or a metal salt thereof; crosslinking agents; colorants or pigments; carbon black; graphite; carbon fibers; and blowing agents; and any and all combinations thereof; and

10 B) said one or more base polymers, Component B, is a homogenous ethylene/C<sub>3</sub>-C<sub>20</sub> alpha-olefin interpolpolymer or a polypropylene or propylene/C<sub>4</sub>-C<sub>20</sub> alpha-olefin copolymer.

5. The polymer composition of Claim 4 wherein;

15 A) said one or more fillers, Component A, is carbon black; alumina trihydrate, calcium carbonate or any and all combinations thereof; and

B) said one or more base polymers, Component B, is a homogenous ethylene/propylene or ethylene/octene-1 copolymer or polypropylene.

6. The polymer composition of Claim 1 wherein

20 i) when calcium carbonate is said filler, the highest ultimate filler loading is greater than 90 weight percent; or

ii) when alumina trihydrate is said filler, the highest ultimate filler loading is greater than 80 weight percent.

25 7. A polymer composition comprising a filler and polymer, selected from homogenous ethylene/C<sub>3</sub>-C<sub>20</sub> alpha-olefin interpolymers or C<sub>3</sub>-C<sub>20</sub> alpha-olefin homopolymers or interpolymers, wherein when said polymer is mixed with calcium carbonate at a concentration of 89 weight percent the viscosity of the resulting mixture (when measured on a Rheometrics RMS-800 with 25 mm parallel plates at frequency of 100 rad/s at 230 °C in a nitrogen purge) is greater than 1.3 x 10<sup>4</sup>  
30 poise.